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Title: Closeout Report: Machine Learned Effective Hamiltonians for Molecular Properties

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Closeout Report:

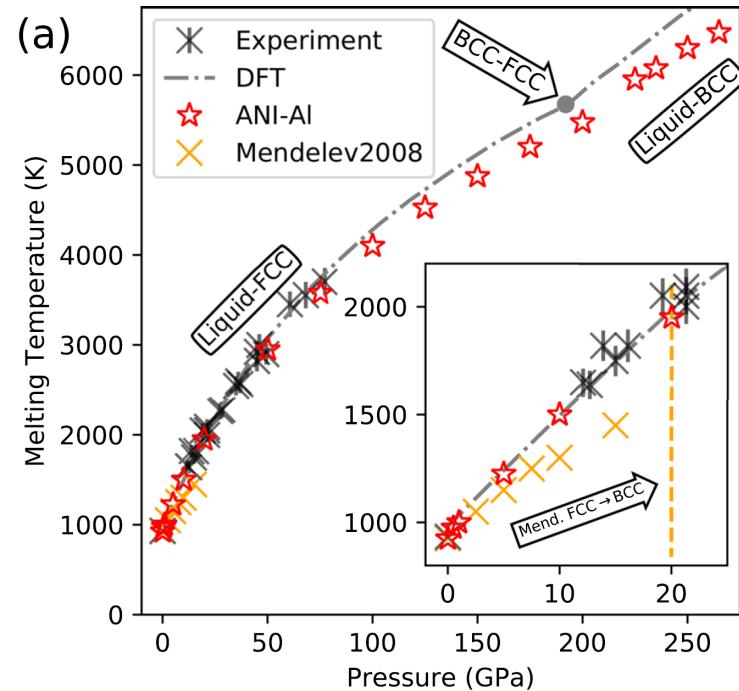
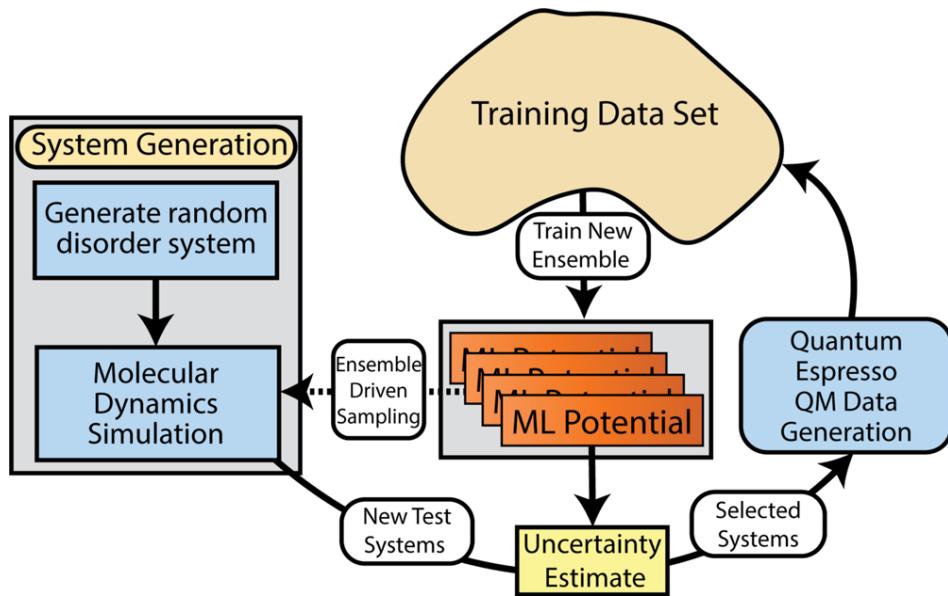
Machine Learned Effective Hamiltonians for Molecular Properties



PI: Ben Nebgen
Co-I: Justin Smith, Sergei Tretiak, Nick Lubbers

2/26/2021

Automated Discovery of Aluminum Potential



An Active Learning system was developed for the automated exploration of phase space (left). This system builds a

Using the dataset produced above, a Machine Learned interatomic potential for Aluminum was developed that closely matches experimental properties (right).

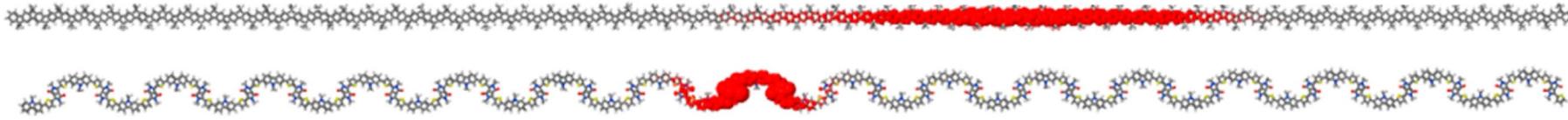
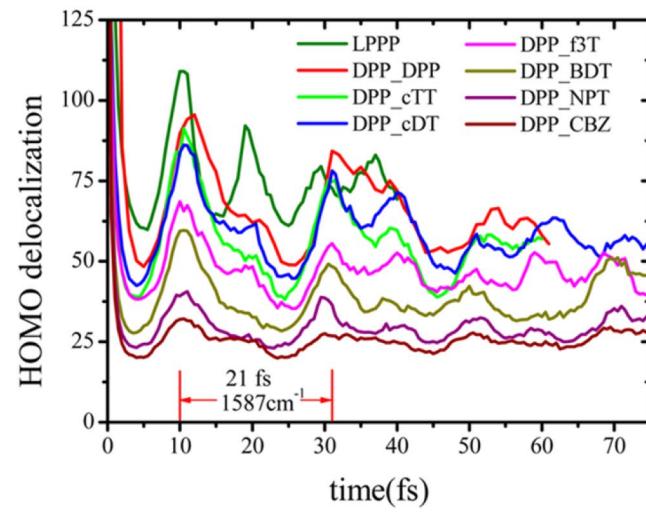
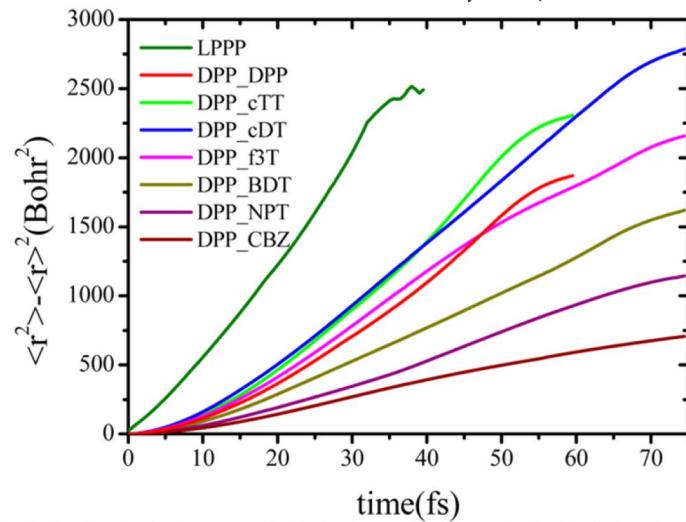
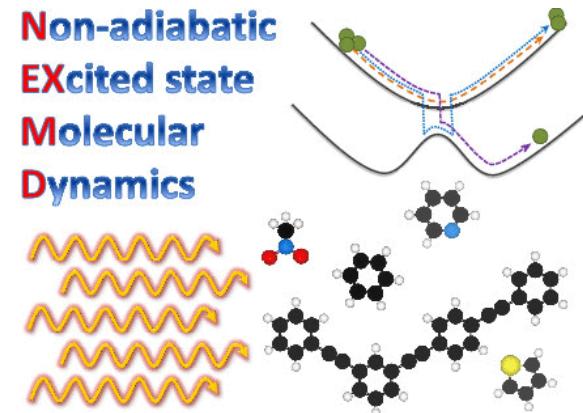
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Development and Release of NEXMD Software

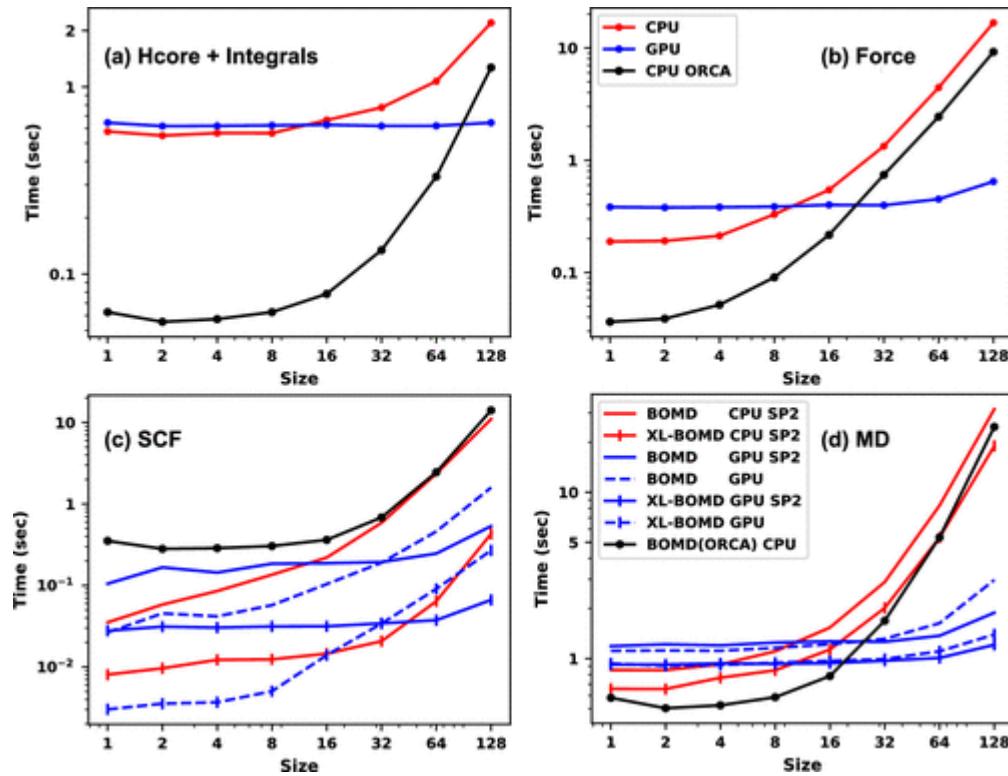
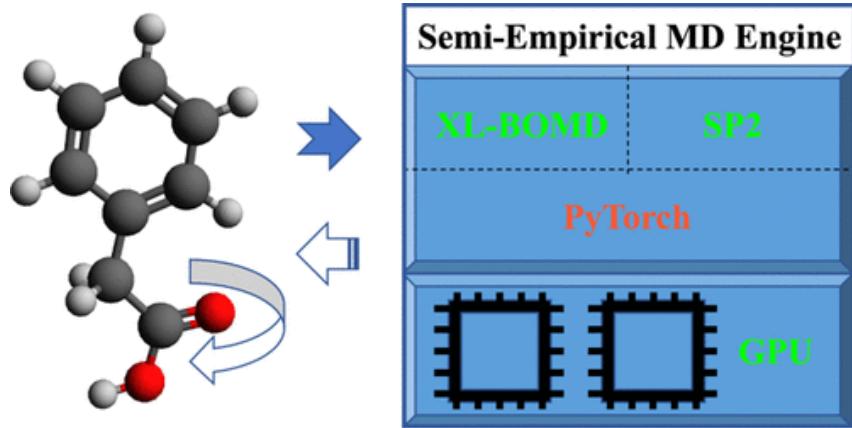
NEXMD is a software used to simulate excited state dynamics of molecular systems, such as charge transport and delocalization in semiconducting polymers (bottom).

Malone, W.; Nebgen, B.; White, A.; Zhang, Y.; Song, H.; Bjorgaard, J. A.; Sifain, A. E.; Rodriguez-Hernandez, B.; Freixas, V. M.; Fernandez-Alberti, S.; Roitberg, A. E.; Nelson, T. R.; Tretiak, S., NEXMD Software Package for Nonadiabatic Excited State Molecular Dynamics Simulations. *J. Chem. Theory Comput.* **2020**, *16*, 5771-5783.

Nelson, T. R.; White, A. J.; Bjorgaard, J. A.; Sifain, A. E.; Zhang, Y.; Nebgen, B.; Fernandez-Alberti, S.; Mozyrsky, D.; Roitberg, A. E.; Tretiak, S., Non-adiabatic Excited-State Molecular Dynamics: Theory and Applications for Modeling Photophysics in Extended Molecular Materials. *Chem Rev* **2020**, *120*, 2215-2287.



Development and Release of PYSEQM Software



Zhou, G.; Nebgen, B.; Lubbers, N.; Malone, W.; Niklasson, A. M. N.; Tretiak, S., Graphics Processing Unit-Accelerated Semiempirical Born Oppenheimer Molecular Dynamics Using PyTorch. *J. Chem. Theory Comput.* **2020**, *16*, 4951-4962.

PYSEQM facilitates semi-empirical quantum mechanical simulations on molecular systems with a Pytorch backend. Calculations can be run on GPU accelerators.

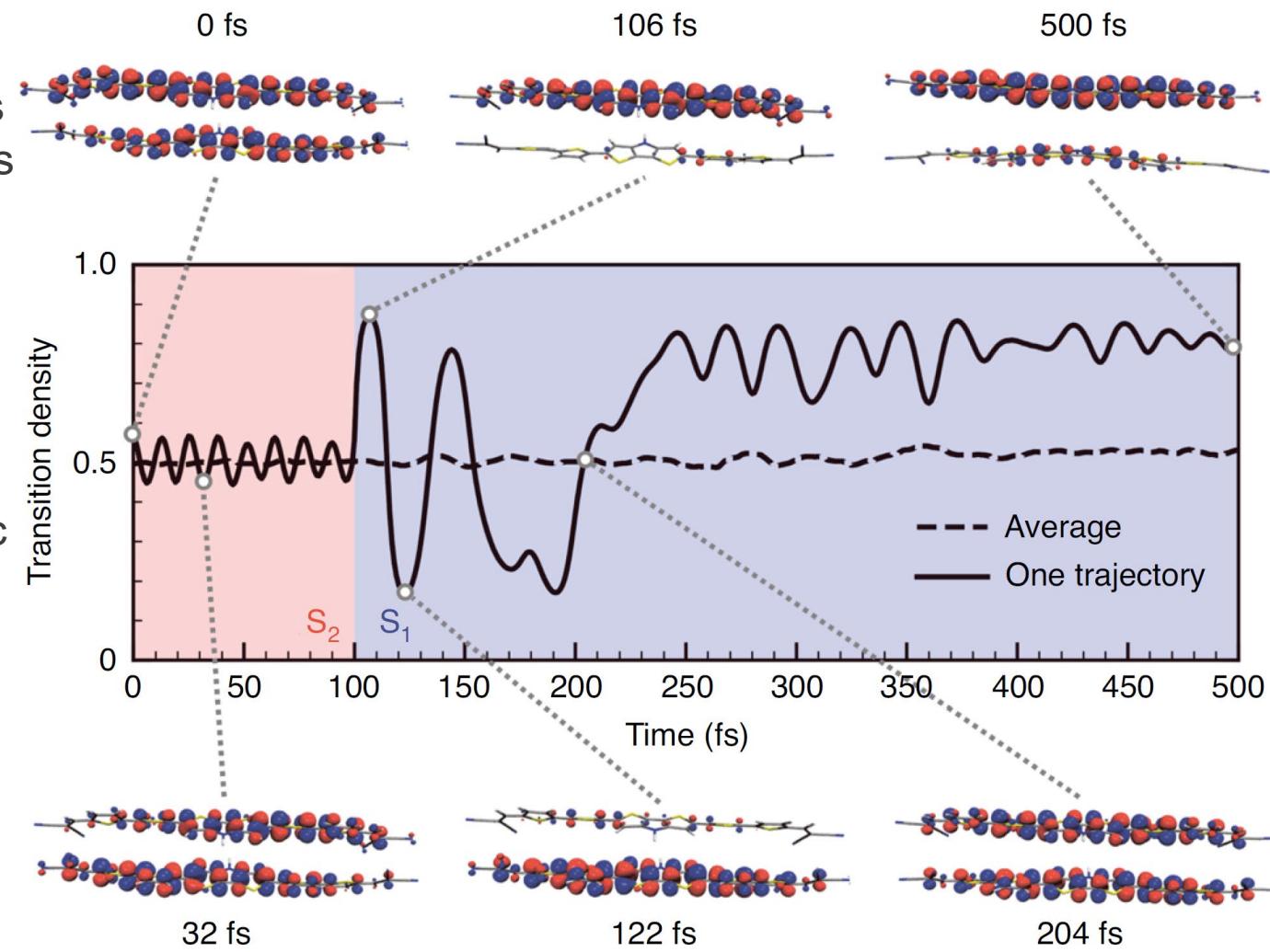
Use of Pytorch allows for interfaces with ML methods for dynamic parameterization of the semi-empirical Hamiltonian

NEXMD Simulations of Interest to Optoelectronics

NEXMD can simulate energy transport and vibrational interactions in candidate molecules for optoelectronic applications.

The results obtained from this study aided the interpretation of ultrafast 2-D electronic spectroscopy.

De Sio, A.; Sommer, E.; Nguyen, X. T.; Groß, L.; Popović, D.; Nebgen, B. T.; Fernandez-Alberti, S.; Pittalis, S.; Rozzi, C. A.; Molinari, E.; Mena-Osteritz, E.; Bäuerle, P.; Frauenheim, T.; Tretiak, S.; Lienau, C., Intermolecular conical intersections in molecular aggregates. *Nature Nanotechnology* **2020**, 16, 63-68.



Public Releases

- Publications
 - Smith, J. S.; Nebgen, B.; Mathew, N.; Chen, J.; Lubbers, N.; Burakovskiy, L.; Tretiak, S.; Nam, H. A.; German, T.; Fensin, S.; Barros, K., Automated discovery of a robust interatomic potential for aluminum. *Nature Comm.* **2021**, 12.
 - De Sio, A.; Sommer, E.; Nguyen, X. T.; Groß, L.; Popović, D.; Nebgen, B. T.; Fernandez-Alberti, S.; Pittalis, S.; Rozzi, C. A.; Molinari, E.; Mena-Osteritz, E.; Bäuerle, P.; Frauenheim, T.; Tretiak, S.; Lienau, C., Intermolecular conical intersections in molecular aggregates. *Nature Nanotechnology* **2020**, 16, 63-68.
 - Malone, W.; Nebgen, B.; White, A.; Zhang, Y.; Song, H.; Bjorgaard, J. A.; Sifain, A. E.; Rodriguez-Hernandez, B.; Freixas, V. M.; Fernandez-Alberti, S.; Roitberg, A. E.; Nelson, T. R.; Tretiak, S., NEXMD Software Package for Nonadiabatic Excited State Molecular Dynamics Simulations. *J. Chem. Theory. Comput.* **2020**, 16, 5771-5783.
 - Nelson, T. R.; White, A. J.; Bjorgaard, J. A.; Sifain, A. E.; Zhang, Y.; Nebgen, B.; Fernandez-Alberti, S.; Mozyrsky, D.; Roitberg, A. E.; Tretiak, S., Non-adiabatic Excited-State Molecular Dynamics: Theory and Applications for Modeling Photophysics in Extended Molecular Materials. *Chem Rev* **2020**, 120, 2215-2287.
 - Zhou, G.; Nebgen, B.; Lubbers, N.; Malone, W.; Niklasson, A. M. N.; Tretiak, S., Graphics Processing Unit-Accelerated Semiempirical Born Oppenheimer Molecular Dynamics Using PyTorch. *J. Chem. Theory. Comput.* **2020**, 16, 4951-4962.
 - Smith, J. S.; Zubatyuk, R.; Nebgen, B.; Lubbers, N.; Barros, K.; Roitberg, A. E.; Isayev, O.; Tretiak, S., The ANI-1ccx and ANI-1x data sets, coupled-cluster and density functional theory properties for molecules. *Sci. Data* **2020**, 7.
- Code Releases
 - NEXMD: <https://github.com/lanl/NEXMD>
 - PYSEQM: <https://github.com/lanl/PYSEQM>